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Surface ocean-lower atmosphere study: Scientific synthesis and contribution to Earth system science

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ABSTRACT

The domain of the surface ocean and lower atmosphere is a complex, highly dynamic component of the Earth system. Better understanding of the physics and biogeochemistry of the air–sea interface and the processes that control the exchange of mass and energy across that boundary define the scope of the Surface Ocean-Lower Atmosphere Study (SOLAS) project. The scientific questions driving SOLAS research, as laid out in the SOLAS Science Plan and Implementation Strategy for the period 2004–2014, are highly challenging, inherently multidisciplinary and broad. During that decade, SOLAS has significantly advanced our knowledge. Discoveries related to the physics of exchange, global trace gas budgets and atmospheric chemistry, the CLAW hypothesis (named after its authors, Charlson, Lovelock, Andreae and Warren), and the influence of nutrients and ocean productivity on important biogeochemical cycles, have substantially changed our views of how the Earth system works and revealed knowledge gaps in our understanding. As such SOLAS has been instrumental in contributing to the International Geosphere–Biosphere Programme (IGBP) mission of identification and assessment of risks posed to society and ecosystems by major changes in the Earth's biological, chemical and physical cycles and processes during the Anthropocene epoch. SOLAS is a bottom-up organization, whose scientific priorities evolve in response to scientific developments and community needs, which has led to the launch of a new 10-year phase. SOLAS (2015–2025) will focus on five core science themes that will provide a scientific basis for understanding and projecting future environmental change and for developing tools to inform societal decision-making.

1. Introduction

In 1990, within the International Geosphere–Biosphere Programme (IGBP) framework for a study of global change (IGBP report No. 12, 1990), the Global Ocean Euphotic Zone Study (GOEZS) was designated as a 'next generation' project to build upon the World Ocean Circulation Experiment (WOCE), the

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Tropical Ocean and Global Atmosphere program (TOGA) and the Joint Global Ocean Flux Study (JGOFS). This new project would integrate their findings and unanswered questions into an interdisciplinary study of the coupled physical, biological, and chemical processes operating in the euphotic zone. In 1993, GOEZS was developing its scientific program as a possible core program of IGBP and the Scientific Committee on Oceanic Research (SCOR) with the support of the World Climate Research Programme (WCRP) (Denman, 1993). GOEZS was formulated as a 'model driven' project, i.e., questions to be studied would be generated from models. Unfortunately, robust predictive models did not exist in the field at that time and so after ample discussions, experts in the field decided that GOEZS would not be established.

Given the importance that IGBP and other organizations placed on the environmental change occurring in the Anthropocene, and the significant influence that ocean interactions have on global environment and society, in 1997 a new project was considered that would cover marine biogeochemistry and its interaction with the atmosphere: the Surface Ocean-Lower Atmosphere Study (SOLAS). It would address key interactions among the marine biogeochemical system, the atmosphere and climate, and how this system affects and is affected by past and future climate and environmental changes. SOLAS was an outgrowth of GOEZS, but was to be based on hypotheses; it would formulate and test hypotheses about key interactions, quantify cause and effect in these interactions, and incorporate this new understanding into models.

Five important hypotheses were identified (Watson, 1997): (i) marine sulfur emissions have a substantial effect on climate by influencing cloud albedo; (ii) atmospherically derived iron stimulates phytoplankton growth in 'high-nitrate-low-chlorophyll' regions of the oceans; (iii) changing patterns of atmospheric nitrogen deposition will significantly influence the marine biota in some parts of the oceans; (iv) the influence of changes in marine biogeochemistry on ocean uptake of anthropogenic carbon dioxide (CO_2) in the next century will be small and (v) the principal effect on marine ecosystems in a warmer world will be a decrease in global productivity, resulting from a slowing of the thermohaline circulation.

In 1999, SOLAS was still in the developmental stage and actively seeking support from the International Global Atmospheric Chemistry (IGAC) and Joint Global Ocean Flux Study (JGOFS) projects, to ensure that atmospheric and oceanic sciences would be properly combined. In 2000, SOLAS moved into an advanced stage of planning by holding an open science meeting in Damp, Germany. Among the 250+ participants were physical, chemical and biological oceanographers, atmospheric chemists and physicists, paleo-oceanographers, remote sensing experts and biogeochemical and climate modelers. The conference provided a platform for these researchers to discuss interdisciplinary collaboration for the first time, to achieve a new scientific understanding of ocean/atmosphere interactions and their susceptibility to perturbation. Stimulating plenary presentations and productive discussions led to the formulation of the overarching questions for SOLAS research and to a draft science plan, which was revised based upon feedback from the community (Wallace, 2000). The document was reviewed in 2003 and after final revisions and approval by SCOR, IGBP, iCACGP (International Commission on Atmospheric Chemistry and Global Pollution), and WCRP it was published in early 2004 (SOLAS, 2004). The SOLAS project had an unusually large number of sponsoring organizations by design, to reflect the highly interdisciplinary nature of the project and bring the oceanographic and atmospheric communities together.

As detailed in the science plan and implementation strategy (SOLAS, 2004), the objective of SOLAS is "to achieve quantitative understanding of the key biogeochemical-physical interactions and feedbacks between the ocean and the atmosphere, and of how this coupled system affects and is affected by climate and

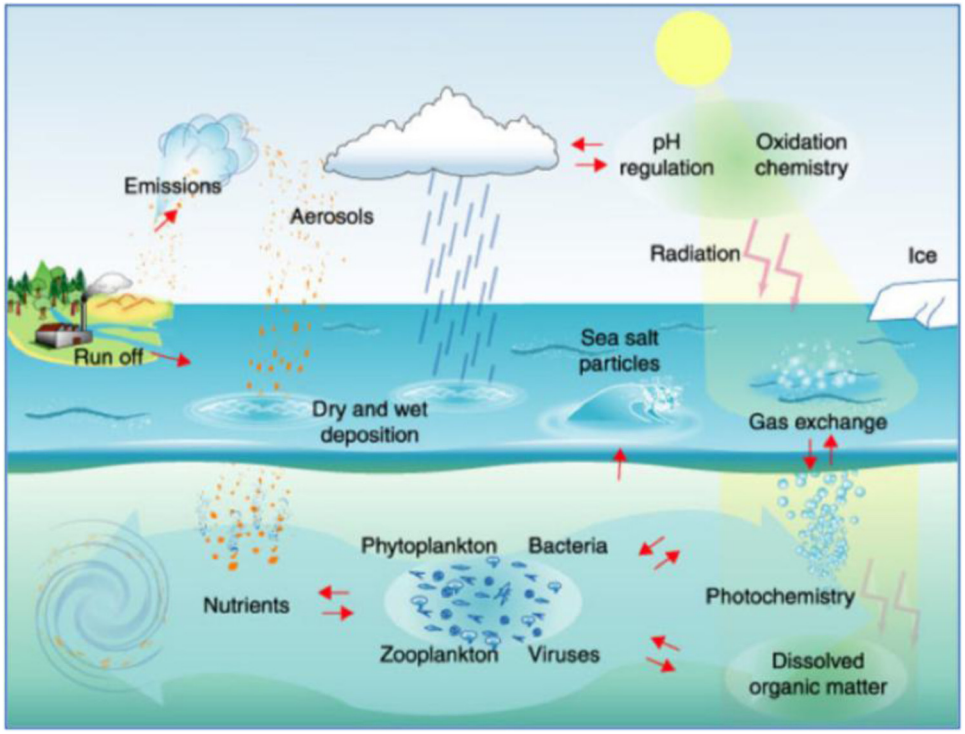

Fig. 1. Diagram to illustrate the domain of SOLAS, its interdisciplinary nature and the main operative processes.
Source: figure from SOLAS (2004).

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environmental change". The interdisciplinary nature and broad domain of SOLAS are illustrated in Fig. 1.

The science plan promoted coupled ocean and atmosphere studies in three focus areas: (i) biogeochemical interactions and feedbacks between the ocean and atmosphere; (ii) exchange processes at the air–sea interface and the role of transport and transformation in the atmospheric and oceanic boundary layers and (iii) air–sea flux of CO_2 and other long-lived radiatively active gases. As predicted in the science plan, new challenges arose in this rapidly evolving field of research that required reassessment of the SOLAS research aims. In 2008 the SOLAS Scientific Steering Committee identified several unresolved issues of significance to the global climate system that would benefit from additional international coordination and networking: upwelling areas and associated oxygen minimum zones, sea ice, marine aerosols, atmospheric nutrient supply and ship emissions (Law et al., 2013).

With continued support from international scientists, the SOLAS project has grown and now encompasses more than 2200 researchers in more than 75 countries. Over the last decade SOLAS has held five open science conferences welcoming over 1250 scientists, six international summer schools training over 420 young scientists, published a textbook based upon the summer school courses (Le Quéré and Saltzman, 2009), sent close to 100 SOLAS e-bulletins, published 15 newsletter issues (http://www.solas-int.org/), had four large national funded programs (Canada, United Kingdom, Germany, Japan) and hundreds of funded SOLAS-related research projects, and orchestrated about one hundred scientific workshops, all of which have underpinned the collaborative community of SOLAS researchers. The International Project Office was hosted from 2003 to 2010 by the University of East Anglia in Norwich, United Kingdom, and since 2010 by the GEOMAR Helmholtz Centre for Ocean Research Kiel in Germany. In 2014, the first phase of SOLAS was completed and marked by the open access publication of a synthesis book (Liss and Johnson, 2014). There have been major advances in our knowledge of ocean–atmosphere exchange processes in the last decade. In the following section, some achievements in major scientific areas are highlighted.

2. Selected major achievements

2.1. Air–sea fluxes

2.1.1. Physics of exchange

One of the goals of the SOLAS program was to reduce uncertainties in air–sea gas exchange because of the importance of this process in the global biogeochemical cycles of many climate-active compounds. Air–sea gas transfer is one of the most challenging problems in environmental science, because of the wide range in scales of mixing near the two-fluid air–sea boundary and the biogeochemical complexity of the air–sea interface. A process-level understanding is required in order to parameterize air–sea gas exchange in a way that accurately captures its coupling to the physical and biogeochemical state of the ocean–atmosphere system.

The SOLAS community carried out multi-investigator survey cruises across the Atlantic and Pacific Oceans, and process studies in the equatorial Eastern Pacific, Southern Ocean, and North Atlantic Oceans (Bell et al., 2013; Ho et al., 2011; Huebert et al., 2010; Marandino et al., 2009; Miller et al., 2009; Yang et al., 2011, 2014). These experiments explored a wide range of conditions from oligotrophic, low wind, stratified tropical waters, to highly mixed, wind-forced, bloom-forming regions of the mid-high latitudes. The cruises involved collaborations between oceanographers, atmospheric scientists, chemists, and physicists. A new generation of chemical sensors was applied to air–sea exchange studies enabling direct flux measurements of climate-active gases (Bariteau et al., 2010; Blomquist et al., 2012; Coburn et al., 2014; Yang et al., 2013), and an array of novel techniques were used to probe the structure, stability, and dynamics of the ocean surface (Pascal et al., 2011; Ward et al., 2014). The observations from these studies challenge existing wind speed-based parameterizations used in the current generation of global biogeochemical models.

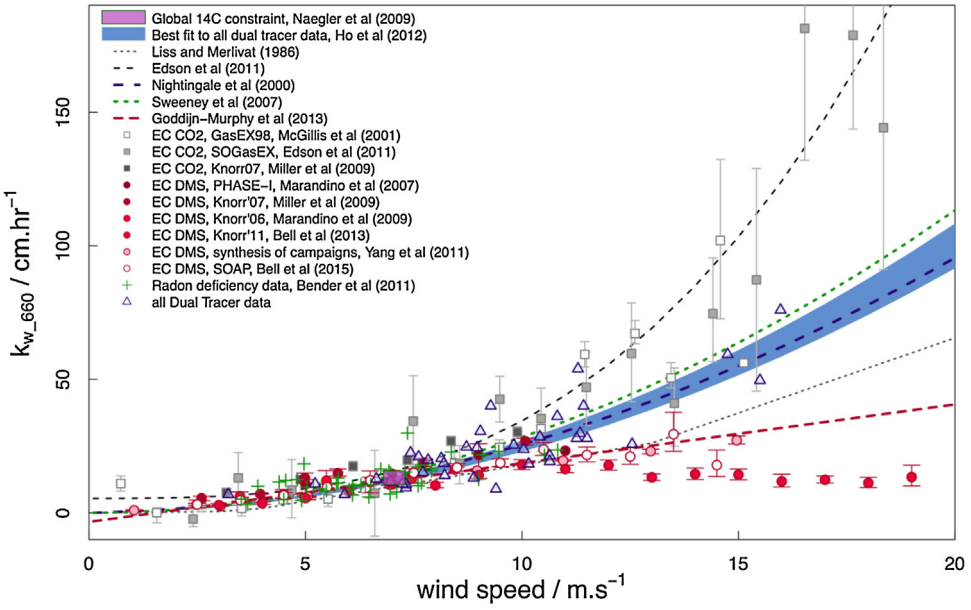

Fig. 2. A comparison of different wind speed relationships of the waterside transfer velocity, k_w. Measurements from eddy covariance techniques and from mass balance techniques are presented. Measurements from eddy covariance techniques and from mass balance techniques are presented (Bell et al., 2013, 2015; Bender et al., 2011; Edson et al., 2011; Goddijn-Murphy et al., 2013; Ho et al., 2012; Liss and Merlivat, 1986; Marandino et al., 2007, 2009; McGillis et al., 2001; Miller et al., 2009; Naegler, 2009; Nightingale et al., 2000; Sweeney et al., 2007; Yang et al., 2011).
Source: figure developed from Johnson (2012); http://dx.doi.org/10.6084/m9.figshare.92419, CC-BY licence.

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An increase in greenhouse gas emissions enhances global atmospheric temperatures, which influence the Earth's pressure gradient and thus wind speed. In addition, 90% of the heat accumulation is in the ocean (IPCC, 2014), causing changes to ocean stratification and circulation. It is important to understand how these changes in the physical forcing of gas exchange may influence air–sea gas transfer in the future. The SOLAS studies demonstrated the feasibility of direct measurement of air–sea gas exchange on time-space scales comparable to the variability in the physical forcings (wind, waves, biologically generated microlayers, etc.). For the first time, air–sea gas fluxes were measured simultaneously with the energy fluxes of sensible heat, latent heat, and momentum. This capability is a step forward to developing physically-based models and bulk parameterizations that predict air–sea fluxes of energy and gases in a self-consistent way (Fairall et al., 2011; Johnson, 2010; Soloviev, 2007) (Fig. 2). Perhaps equally important, the SOLAS project (field studies, summer schools, workshops, and open science conferences) helped build a community of young scientists engaged in air–sea exchange research with expertise crossing the traditional boundaries of atmospheric and oceanic sciences.

2.1.2. Global fluxes: the Surface Ocean CO_2 Atlas

The global oceans constitute an important net sink for the greenhouse gas carbon dioxide (CO_2) (Takahashi et al., 2002). The concept that the ocean is the largest sink for anthropogenic CO_2, but that the air–sea flux of CO_2 may be changing, was one of the five hypotheses driving SOLAS and one of the three foci of the science plan and implementation strategy (SOLAS, 2004). Accurate knowledge of the surface water CO_2 distribution, in combination with the air–sea gas transfer velocity (Fig. 2), enables quantification of the size of this important sink. Hence, systematic, high-quality CO_2 measurements, data reporting and data synthesis are essential. To this end the international marine carbon research community initiated the Surface Ocean CO_2 Atlas (SOCAT) in 2007 (IOCCP, 2007). SOCAT makes surface water CO_2 data available through regular releases of quality controlled and documented, synthesis f_{CO2} (fugacity of CO_2) products for the global ocean and coastal seas (Bakker et al., 2012, 2014a; Pfeil et al., 2012; Sabine et al., 2013). SOCAT version 1 was released in 2011, followed by version 2 in 2013, and version 3 in 2015. The SOCAT data products are available for download from www.socat.info, where they are archived and can also be used interactively.

Version 3 contains 14.5 million surface water fCO_2 values from 1968 to 2014 (Bakker et al., 2014a, in preparation). They originate from seagoing fieldwork by scientists in 22 countries. The data were collected on more than one hundred ships, moorings and drifters. They are submitted to the database by individual scientists and quality control is then carried out by volunteer scientists prior to release. The increase in data collection over the past four decades is striking (Fig. 3). Installation of autonomous, infrared CO_2 instruments on ships has provided repeated fCO_2 observations along major shipping lines from the early 1990s onwards. Nonetheless, the observations are sparse for much of the world's oceans.

Numerous peer-reviewed, scientific publications and high-profile reports cite SOCAT (www.socat.info). Applications of SOCAT include: quantification of the ocean carbon sink and its variation

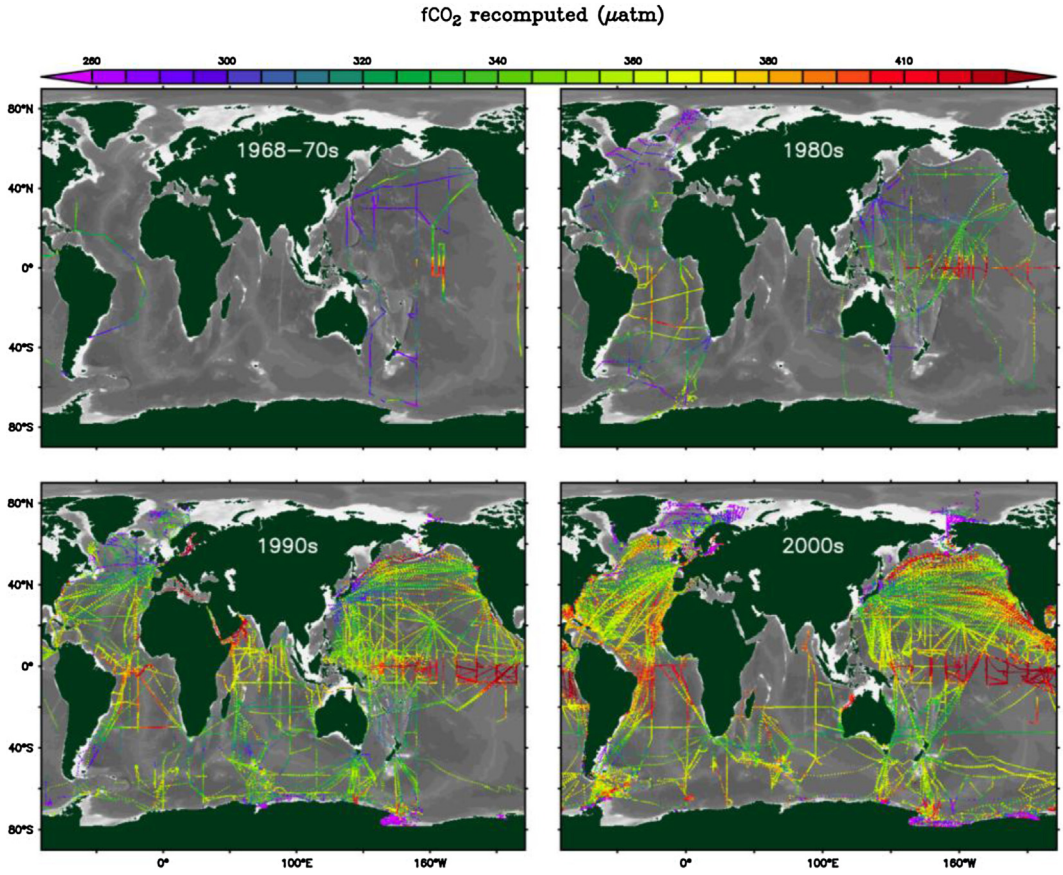

Fig. 3. Surface water fCO_2 values in 1968 to 1979, the 1980s, 1990s and 2000s in the global ocean and coastal seas for SOCAT version 2. Pfeil et al. (2012) present a similar figure for version 1.
Source: figure prepared with the Cruise Data Viewer at www.socat.info.

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(e.g., Le Quéré et al., 2014); provision of constraints for atmospheric inverse models used for estimating the land carbon sink; validation of ocean carbon models; studies of ocean acidification, ocean carbon cycling and genomics. Sustained, long-term surface ocean CO_2 observations and their synthesis are critical for early detection of any changes in atmospheric carbon fluxes.

Revised quality control criteria will enable inclusion of well-calibrated CO_2 measurements by alternative sensors and on alternative platforms from version 3 onwards (Wanninkhof et al., 2013). For its version 4, SOCAT will accept, archive and make public additional parameters accompanying surface water CO_2 data, such as nutrients (SOCAT, 2014). SOCAT will not quality control the additional parameters, but would welcome other synthesis activities to do so. An automated data upload is available and allows preliminary quality control during data submission. As a consequence, it should enable a more rapid release of SOCAT data products.

2.1.3. Distribution and fluxes of nitrous oxide and halocarbons

Both nitrous oxide (N_2O) and halocarbons have received much attention within the SOLAS community; they are featured in two of the three foci of the SOLAS science plan and implementation strategy (SOLAS, 2004). N_2O is an atmospheric trace gas that plays an important role in both atmospheric chemistry and Earth's climate. The ocean is a major natural source of atmospheric N_2O (IPCC, 2014), but global oceanic emission estimates are still associated with a high degree of uncertainty. This is partly caused by the fact that there was no database which could provide global oceanic N_2O data sets. To this end, MEMENTO (the MarinE MethanE and NiTrous Oxide database; https://memento.geomar.de/de) was initiated by the European CoOperation in Science and Technology framework (COST) Action 735 and SOLAS (Bange et al., 2009). COST Action 735 (2006–2011) aimed to develop tools for assessing global air–sea fluxes of climate and air pollution relevant gases. Since 2014, MEMENTO is closely working with the SCOR working group 143 (https://portal.geomar.de/web/scor-wg-143). Additionally, several new aspects of the oceanic biogeochemistry and the air–sea exchange of N_2O, as well as new analytical methods, have emerged during the SOLAS period, leading to a fundamental change in our understanding of oceanic N_2O (Bakker et al., 2014b): (i) the long-standing paradigm of a predominant bacterial nitrification of N_2O has been challenged by the fact that N_2O is mainly produced by nitrifying archaea (Löscher et al., 2012; Santoro et al., 2011); (ii) a study in the eastern North Atlantic Ocean points to an underestimated role of surfactants in suppressing air–sea gas exchange of N_2O in areas of high biological productivity (Kock et al., 2012); (iii) a model study revealed that the effect of atmospheric nitrogen deposition on oceanic N_2O production is small on a global scale but could be significant on a regional scale (e.g., in the Arabian Sea) (Suntharalingam et al., 2012); (iv) the development of laser-based spectrometers using the cavity-ringdown approach coupled to an equilibrator allows N_2O measurements in surface waters with an unprecedented high temporal and spatial resolution (Arévalo-Martínez et al., 2013; Grefe and Kaiser, 2014) and (v) the first measurements of N_2O in sea ice lead to a new appraisal of N_2O ocean atmosphere fluxes during ice formation and decay (Randall et al., 2012). Future projections of N_2O production in the ocean and subsequent emission to the atmosphere are related to enhancements of so-called oxygen minimum zones (OMZs). OMZs are thought to be expanding due to anthropogenic activities and it has been observed that the nitrogen cycle is perturbed therein, producing large quantities of N_2O as a byproduct (Arévalo-Martínez et al., 2015). Therefore, N_2O production in OMZs may have a positive feedback on global change since it is also a powerful greenhouse gas that alters climate. However, a recent model study showed that the future overall oceanic N_2O emissions might decrease mainly because of an increasing storage capacity (i.e., reduced ventilation) of N_2O in the future ocean (Martinez-Rey et al., 2015). Therefore, the future development of the oceanic N_2O emissions is still under debate.

Halogenated organic compounds from the ocean contribute to the pool of reactive atmospheric halogens. They are involved in ozone depletion in the troposphere and stratosphere and influence aerosol formation. Interestingly, in the case of iodine, marine boundary layer concentrations were thought to be prohibitively low until a recent set of analytical advances have demonstrated that atmospheric iodine chemistry is widespread (Liss and Johnson, 2014 and references therein). Model and laboratory studies now show that atmospheric iodine chemistry results in new particle formation and shifts in the hydrogen oxide radicals (HO_x) ratio and nitrogen oxide radicals (NO_x) ratios, which highly influence the oxidative capacity of the atmosphere (Plane et al., 2006; Saiz-Lopez et al., 2012). During SOLAS, the oceanic source strengths and biogeochemical cycling of iodinated, brominated and chlorinated halocarbons have been investigated. The sparse database of halocarbons in ocean and atmosphere increased considerably during cruises into various oceanic regions at all latitudes during the last decade (e.g., Brinckmann et al., 2012; Butler et al., 2007; Liu et al., 2011; O'Brien et al., 2009; Pyle et al., 2011; Yokouchi et al., 2008). Much of these data has been compiled in the Halocarbons in the Ocean and Atmosphere (HalOcAt) database project (https://halocat.geomar.de/), which is still ongoing and currently consists of 200 data sets, comprising roughly 55,000 oceanic and 470,000 atmospheric data points of 19 different short-lived halogenated compounds. The first comprehensive global sea-to-air flux climatologies of the three important short-lived halogen carriers bromoform ($CHBr_3$), dibromomethane (CH_2Br_2) and methyl iodide (CH_3I) have been derived using the HalOcAt database (Ziska et al., 2013). The impact of these emissions on stratospheric ozone depletion was found to be highly dependent on the magnitude, location, and timing of their emission, being particularly significant in the tropics, but impacting the entire global atmosphere (Hossaini et al., 2013; Liang et al., 2010; Ordóñez et al., 2012; Tegtmeier et al., 2012). Novel process studies in the natural environment and modeling have started to further unravel the pathways of abiotic and biotic production and degradation mechanisms of the halocarbons in the current and future ocean (Hense and Quack, 2009; Hopkins et al., 2013; Hughes et al., 2013; Shi et al., 2014). The influence of meteorological constraints on the air–sea exchange of halocarbons has been investigated (Fuhlbrügge et al., 2013) and new tools developed to determine their source distribution (Ashfold et al., 2014). Recent reviews (Carpenter et al., 2012; Liss et al., 2014) call for a quantification of the relative roles of, and interactions between, the oceanic production and temporal variations of physical forcings, in conjunction with anthropogenic influences as oceanic halocarbon emissions will likely increase in the future (Hepach et al., 2014).

2.2. Evolution of the CLAW hypothesis

In the 1980s, it was hypothesized that dimethylsulfide (DMS)-derived sulfate made up the majority of cloud condensation nuclei (CCN) in the remote marine boundary layer (MBL) (Charlson et al., 1987; Shaw, 1983). Charlson et al. (1987) further hypothesized that an increase in DMS emission from the ocean would result in an increase in CCN, cloud droplet number concentration, and cloud albedo, as well as a decrease in the amount of solar radiation reaching Earth's surface. The reduction in solar radiation would then result in changes in the speciation and abundance of phytoplankton that produce dimethylsulfoniopropionate (DMSP),

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the precursor of DMS, thus setting up a climate feedback loop between cloud albedo and surface ocean DMS concentration. This proposed mechanism for biological regulation of climate became known as the CLAW hypothesis, named after the four authors of Charlson et al. (1987; Charlson, Lovelock, Andreae and Warren).

The climate feedback loop proposed by Charlson et al. requires (1) that a change in the emission of DMS results in a significant change in MBL CCN concentration, (2) a change in DMS-derived CCN yields a change in cloud albedo, and (3) a change in cloud albedo, surface temperature, and/or incident radiation leads to a change in DMS production. The CLAW hypothesis spawned over 25 years of interdisciplinary research, with the biological oceanography, atmospheric chemistry, and climate modeling communities working together to assess the response to change in each of these three steps.

The CLAW hypothesis was one of the main driving hypotheses formulating the SOLAS mission. It outlines a clear biogeochemical interaction and feedback between the ocean and atmosphere that resides at the core of SOLAS research. During the SOLAS decade, more than 500 DMS-related studies accomplished within the broader SOLAS community have been published contributing more than half of the data in the Global Surface Water DMS Database (http://saga.pmel.noaa.gov/dms/). New insights into surface ocean biological production pathways and cycling, such as hypotheses explaining DMS(P) biogenic production (Stefels et al., 2007) and the so-called summer paradox (Vallina et al., 2008) were made. Technological advancements in direct measurement of open ocean DMS fluxes have resulted from SOLAS initiatives (e.g., Huebert et al., 2004; Marandino et al., 2007). The importance of chemical compounds other than the hydroxyl radical, OH, in DMS oxidation reactions (e.g., BrO) has also been identified (Breider et al., 2010; Lawler et al., 2009). A major accomplishment by the combined efforts of SOLAS and COST Action 735 has been to update the first DMS climatology from Kettle et al. (1999), Kettle and Andreae (2000). The subsequent climatology by Lana et al. (2011) gives a more robust calculation of the seasonal, global DMS oceanic concentrations and air–sea fluxes based on the enhanced database. It has been and will continue to be used to better model the effects of DMS emissions on atmospheric chemistry and climate (Levasseur, 2013).

In the years since CLAW was first proposed, new SOLAS observations have become available that complicate the three steps in the simple feedback loop proposed. For example, observations based on direct and indirect chemical techniques have revealed that up to half of the particles in the CCN size range contain sea salt (Campuzano-Jost et al., 2003; Murphy et al., 1998; O'Dowd and Smith, 1993). In addition, measurements below stratocumulus clouds over remote ocean regions have revealed that the majority of residual particles from evaporated cloud droplets – that is, particles that had acted as CCN – were sea salt (Twohy and Anderson, 2008). These measurements, as well as others carried out over the past several decades, show that sea salt can make up a significant fraction of MBL CCN. In addition, the importance of organic-containing particles as CCN has also been revealed over the past decade. Breaking surface ocean waves result in the entrainment of air bubbles that scavenge organic matter from seawater as they rise to the surface. When injected to the atmosphere, the bubbles burst and yield submicrometer sea spray aerosol (Bates et al., 2012; Facchini et al., 2008; Keene et al., 2007; O'Dowd et al., 2004; Quinn et al., 2014). These organic particles, containing surface active gel- forming lipo-polysaccarides, are debated in the literature concerning their hygroscopic and CCN properties (Facchini et al., 2008; Leck and Bigg, 2008; Orellana et al., 2011; Ovadnevaite et al., 2011; Prather et al., 2013; Quinn et al., 2014; Russell et al., 2010). The large contribution of wind-driven sea spray containing both sea salt and organics to the MBL CCN population prevents a significant response in CCN concentration to changes in the emission of DMS.

Furthermore, observations of particle nucleation involving sulfuric acid, a DMS oxidation product, in the free troposphere

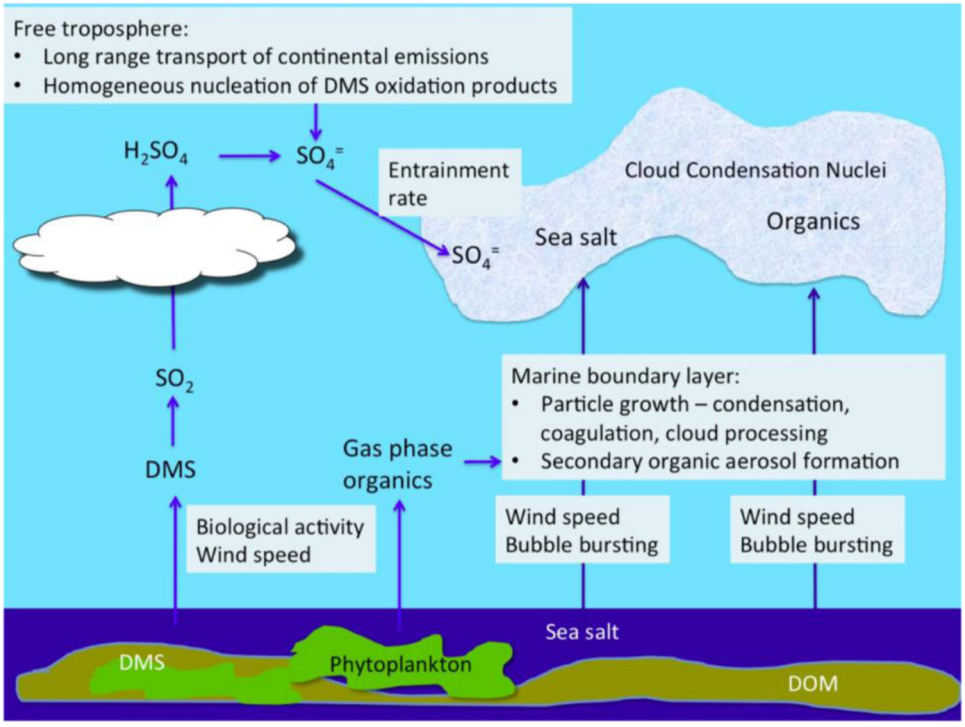

Fig. 4. Sources and production mechanisms for CCN in the remote MBL. DMS contributes to the MBL CCN population primarily via particle formation in the free troposphere in cloud outflow regions with subsequent subsidence. Sea salt and organics are emitted as a result of wind driven bubble bursting.
Source: figure adapted from Quinn and Bates (2011).

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near cloud top height rather than in the MBL are numerous (e.g., Clarke et al., 1998). Gases, including DMS, and particles are mixed from the MBL into clouds. Clouds scavenge the particles, but not insoluble gases, so that the air detrained from the cloud contains low aerosol surface area. In these cloud outflow regions, where existing particle surface area is low, and water vapor concentrations and light levels are high, DMS can undergo gas to particle conversion. Measurements and model calculations published since the introduction of the CLAW hypothesis indicate that DMS-derived sulfate contributes generally to the MBL CCN population via particle nucleation in the free troposphere rather than in the MBL. After formation in the free troposphere, the particles may be transported thousands of kilometers before mixing down into the MBL. As a result, regions of high DMS seawater-to-air fluxes may not always correlate with regions of high DMS-derived CCN concentrations.

Field and laboratory experiments combined with model calculations performed over the past decades have shown that sources of CCN to the MBL are more complex than was recognized by the CLAW hypothesis. The concentration of CCN in the MBL is a result of emissions of sea salt and organics in sea spray, sinking of DMS- and continentally-derived particulates from the free

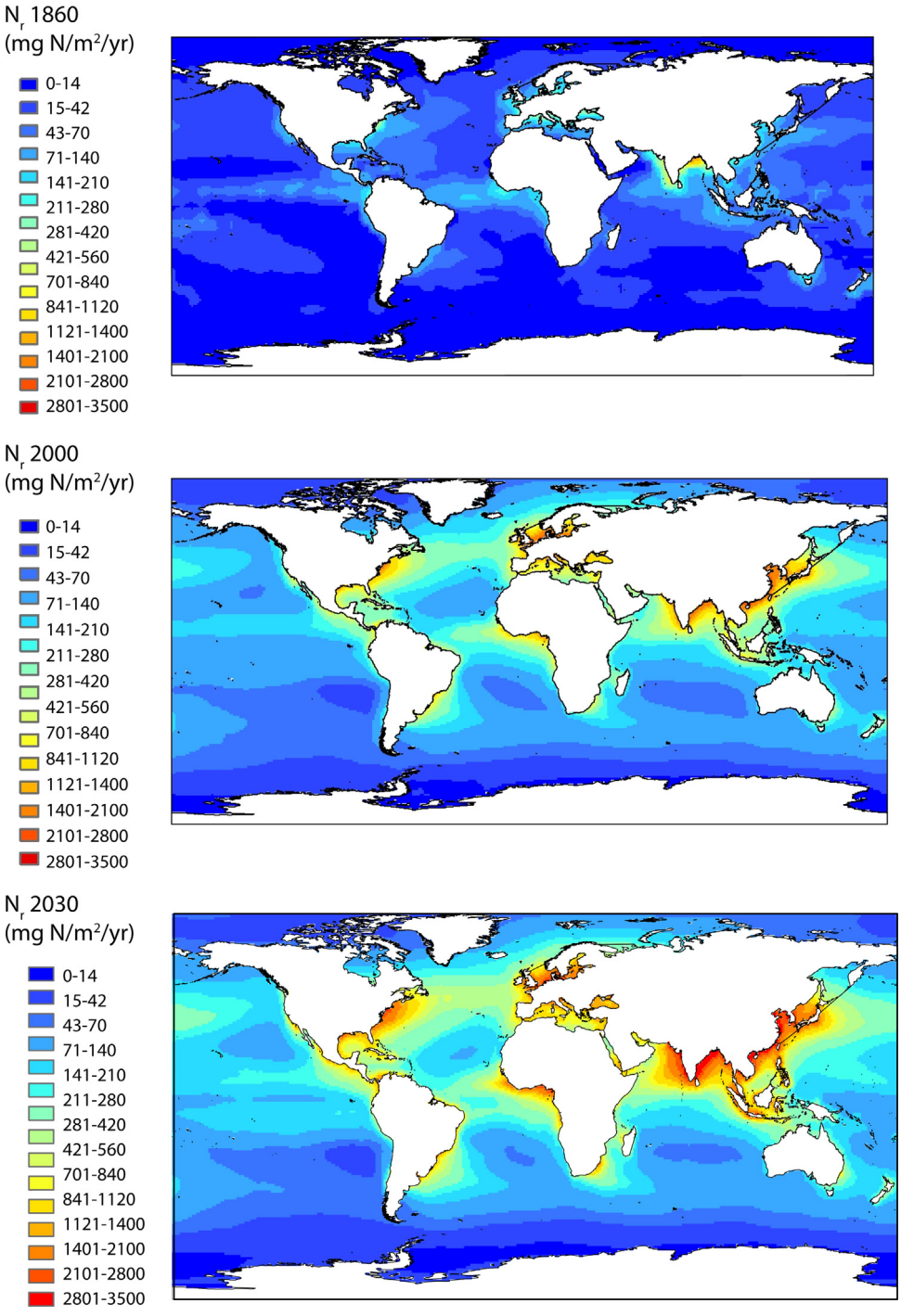

Fig. 5. Total atmospheric reactive nitrogen (Nr) deposition to the ocean in mg m^{-2} year^{-1} in 1860, 2000, and 2030. Both inorganic and organic forms of nitrogen are included. Source: figure adapted from Duce et al. (2008).

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troposphere, and particle growth through coagulation, vapor condensation, and cloud processing (Fig. 4). Although the CLAW hypothesis in its original formulation has not stood the test of time, it was a revolutionary paradigm in Earth system science and charted the way for the interdisciplinary research now required to fully understand the multiple sources and climate impacts of remote MBL CCN (Quinn and Bates, 2011).

2.3. Nutrients and ocean productivity

2.3.1. Nitrogen deposition

Nitrogen limits primary production in large areas of the ocean. Most marine organisms utilize oxidized and reduced inorganic and organic forms of fixed or reactive nitrogen, Nr. The three open ocean sources of external (not recycled) Nr are biological N_2 fixation, riverine input, and atmospheric deposition. External sources contribute a net oceanic input of Nr that support "new production". Changes in the relative importance of these external sources influence global oceanic Nr, carbon sequestration, and affect CO_2 air–sea exchange.

In 2006, SOLAS and the International Nitrogen Initiative convened a workshop on "Anthropogenic Nitrogen Impacts on the Open Ocean" that evaluated anthropogenic atmospheric nitrogen entering the ocean and its impact on marine biological productivity and possible CO_2 drawdown (Duce et al., 2008). They found that a significant fraction of the global emissions of atmospheric nitrogen species deposit on the ocean surface. While most was inorganic nitrogen (nitrate and ammonium), ~30% was water-soluble organic nitrogen, which had not been considered previously in global models (Ito et al., 2015; Kanakidou et al., 2012). Duce et al. (2008) showed that in 2000 these increasing quantities of atmospheric Nr entering the open ocean may have accounted for ~1/3 of the ocean's external nitrogen supply, and up to ~3% of the annual new marine biological production, representing a few percent of the ocean's drawdown of CO_2. Others SOLAS-related studies (Krishnamurthy et al., 2009, 2010) demonstrated that increasing nitrogen inputs alone increased small phytoplankton and diatom production, leading to phosphorous and iron limitation of diazotrophs and reducing nitrogen fixation.

Fig. 5 (top and middle) shows that there have been significant changes in the spatial distribution of marine Nr deposition since 1860. By 2000 strong plumes of deposition extended far downwind of many major urban areas. Estimates for 2030 (bottom) suggest Nr oceanic deposition will be four times that in 2000 (Duce et al., 2008). If so, atmospheric anthropogenic nitrogen contributions to marine primary production could approach current estimates of global marine N_2 fixation. Increases in the surface nitrate concentration in the northwest Pacific were recently documented, indicating atmospheric transport of anthropogenic nitrogen from Asia (Kim et al., 2014a). Studies have also shown that areas in the northern Indian Ocean (Singh et al., 2012; Srinivas and Sarin, 2013), the South China Sea, northwest Pacific (Jung et al., 2013; Kim et al., 2011, 2014b; Uematsu et al., 2010) and the North Atlantic (Baker et al., 2010; Lesworth et al., 2010) are now being impacted by atmospheric nitrogen deposition.

Several studies provide new perspectives on this issue. Using a multi-model approach to evaluate the mean nitrogen deposition to continental and oceanic areas for the present, for 2030 and 2100, Lamarque et al. (2013) suggested there will be decreasing deposition of oxidized Nr later in this century, reflecting the anticipated improvement in nitrogen oxide radicals emission control, while ammonia deposition will continue to increase. However, using nitrogen isotope data and marine versus continental back trajectory analysis, most ammonia deposition at Bermuda was found to be marine derived, not anthropogenic (Altieri et al., 2014). If this occurs in other marine regions, many of the earlier estimates of anthropogenic input of reduced inorganic nitrogen will have been too high. In addition, it appears that deposition of Nr to low nutrient, low chlorophyll regions was underestimated by models on daily to weekly timescales because models typically overlook large synoptic variations in atmospheric nutrient deposition (Guieu et al., 2014). There is clearly still much work to be done to accurately assess the impact of anthropogenic nitrogen deposition to the ocean.

2.3.2. Iron biogeochemistry and the Iron Addition Experiments

Over the last fifteen years, multi-faceted research into oceanic iron, encompassing regional distributions, sources and sinks, biological recycling, 'paleo iron', and stable isotopes, has evolved into the integrative discipline of iron biogeochemistry (Boyd and Ellwood, 2010). SOLAS scientists have been instrumental in characterizing a wide range of aerosol particles, including desert dust, pollutants, volcanic ash, and their modes of transport, interaction, temporal and spatial signatures, and iron solubility

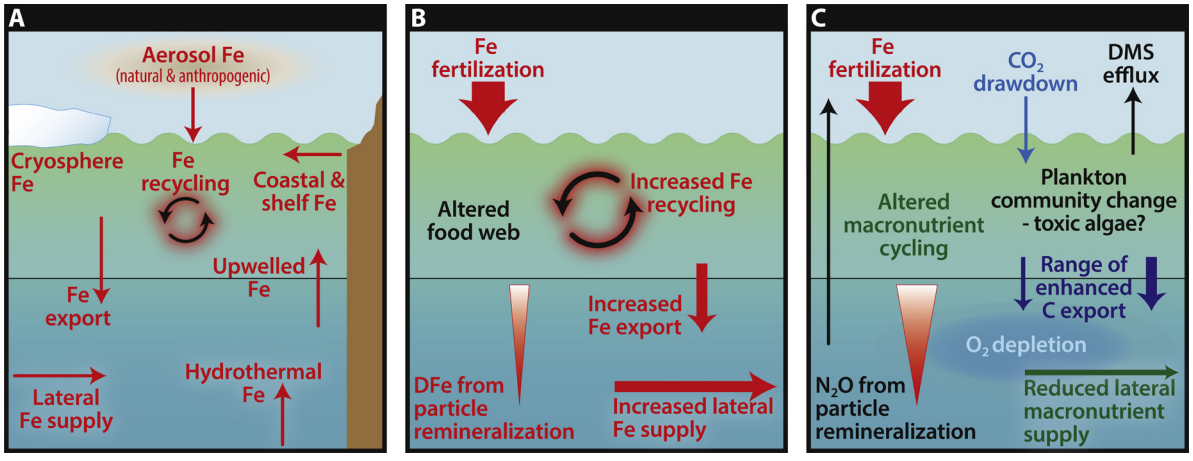

Fig. 6. The oceans biogeochemical iron (Fe) cycle and its ramifications for global climate. (A) Fe supply mechanisms from the atmosphere and ocean, and oceanic biological recycling; (B) implications of deliberate iron fertilization for the natural Fe cycle; and (C) potential implications of deliberate iron fertilization for climate related processes. Solid horizontal line denotes sunlit zone of surface mixing. The two blue downward arrows in (C) represent the known range of export efficiencies estimates from <10% (Boyd et al., 2004) to ~50% (Smetacek et al., 2012) from FeAXs. The downward pink triangles (panels B and C) represent particle attenuation due to biological remineralisation with depth.
Source: figure designed by Hilarie Cutler/IGBP.

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(Baker and Croot, 2010; Landing and Paytan 2010; de Leeuw et al., 2014), which led to the recent development of a SOLAS aerosol and rainwater chemistry database (http://tinyurl.com/aerosol-rainwater) (Garçon et al., 2014). These studies have also largely resolved the enigma of widely differing estimates of aerosol iron solubility (Jickells and Spokes, 2001), and quantified the relative contribution of aerosols to oceanic iron supply and productivity (Boyd et al., 2010), the influence of Asian dust on the global radiation budget (Uno et al., 2009), and the stimulation of upper ocean carbon export resulting from natural iron deposition following volcanic eruptions (Achterberg et al., 2013; Hamme et al., 2010). These advances in our understanding of atmospheric links with the ocean iron cycle have improved modeling parameterizations and estimates of atmospheric dust supply to the ocean iron biogeochemical cycle on past, present and future timescales (Mahowald et al., 2009).

SOLAS field research campaigns examining iron biogeochemistry have ranged from repeat transects that have established the spatial gradients and temporal trends in aerosol deposition (Baker et al., 2006), to studies of upper ocean iron cycling. New insights into iron biogeochemistry have been obtained from detailed pelagic iron budgets; for example, the FeCycle quasi-lagrangian study identified that microbes in High Nutrient Low Chlorophyll (HNLC) waters obtain 90% of their iron requirement from recycling, and consequently that iron is rapidly cycled over timescales of hours (Boyd et al., 2005b). Further research has examined the role of different ligands in maintaining iron availability in surface waters (Hassler et al., 2011). However, iron is not only important in HNLC waters, but is also a critical co-limiting nutrient of productivity in other regions (Moore et al., 2013). These advances in our understanding of iron biogeochemistry (see Fig. 6A) are reflected in the increased representation of iron parameters (stocks, rates and processes) in biogeochemical models (Tagliabue et al., 2010).

Since the inception of SOLAS, the emerging technique of in-situ manipulation experiments has evolved within a number of ocean Iron Addition Experiments (FeAXs), including the multi-platform Canadian-SOLAS SERIES experiment and the repeat Japanese SEEDS FeAX. SERIES produced detailed models of biogenic gas (DMS) production and the first carbon budget relating CO_2 drawdown to iron-stimulated carbon export below the permanent pycnocline (Boyd et al., 2004; Le Clainche et al., 2006; see Fig. 6C). SEEDS showed striking differences in response between years at the same location, largely due to interannual variability in initial oceanic conditions and plankton seed stock composition (Tsuda et al., 2007). Following the first FeAX intercomparison of biogeochemical responses (de Baar et al., 2005), a SOLAS co-sponsored workshop synthesized the findings from 12 FeAXs, and compared the outcomes with those of naturally high iron regions (KEOPS and CROZEX voyages; Blain et al., 2007; Pollard et al., 2009), as well as dust input during episodic events and on glacial timescales (Boyd et al., 2007). The FeAXs were successful in establishing that iron availability controls primary productivity and influences carbon export in HNLC regions (Boyd et al., 2004; Smetacek et al., 2012), and the observational data from the FeAXs were synthesized by the SCOR Working Group "The legacy of mesoscale ocean enrichment experiments" (Boyd et al., 2012). An unanticipated outcome of the FeAXs, arising from the stimulation of phytoplankton growth by iron addition, was interest in deploying iron addition at large-scales to mitigate CO_2 emissions. Subsequent analysis suggested that iron was less effective in enhancing carbon export (Boyd and Browman, 2008; Boyd et al., 2005a). SOLAS issued a position statement and subsequently advised the Intergovernmental Oceanic Committee (IOC) on amendment of the London Convention on Marine Dumping (LC/LP, 2013) to incorporate regulation of iron addition to the ocean. SOLAS has also produced a summary for policy makers for IOC/UNESCO (Wallace et al., 2010), with an associated synthesis paper examining the pros and cons of iron addition for CO_2 mitigation (Williamson et al., 2012; see Fig. 6C). This issue of ocean iron fertilization is one example of the SOLAS commitment to robust scientific underpinning of policy and legislation, and the socioeconomic relevance of ocean–atmosphere research.

3. SOLAS links with Earth system science and IGBP

For almost half a century, it has been established that planetary cycles, such as the hydrological and carbon cycles, are closely interlinked. In fact, life itself is an active and necessary player in these planetary dynamics, as presented by Lovelock with the Gaia hypothesis (Lovelock and Margulis, 1974). The sum of our planet's interacting physical, chemical, and biological processes represents the 'Earth system', in which the ocean, atmosphere and land, as well as the living and non-living parts therein, are all connected. Twenty years ago, the understanding of how the Earth worked as a system, how the components of the system were connected, or even the importance of the individual components, were in their infancy. Feedback mechanisms were more elusive than at present, as were the dynamics controlling the coupled system (Steffen et al., 2004). Earth system science is now at the core of IGBP, which is structured around three major compartments (land, ocean and atmosphere) (IGBP, 2006). Presented here are just some of many findings from SOLAS science that have substantially changed our views of how the Earth system works but revealed gaping holes in our understanding. They hint at the importance of the ocean–atmosphere interface in terms of buffering or accelerating changes in the Earth system. More details of these and other advances are available in the open access book 'Ocean-Atmosphere Interactions of Gases and Particles' by Liss and Johnson (2014).

The IGBP vision is to provide scientific knowledge to improve the sustainability of the living Earth (IGBP, 2006). To this end, in the late 2000s, the novel concept of planetary boundaries emerged to inform societal decisions about sustainability. Nine planetary boundaries within which humanity can continue to develop and thrive for generations to come have been identified and control variables have been quantified. Crossing these boundaries could generate abrupt or irreversible environmental changes; conversely, respecting the boundaries reduces the risks to human society of crossing these thresholds (Rockström et al., 2009; Steffen et al., 2015). The nine boundaries are climate change, biodiversity integrity, biogeochemical flows (P and N cycles), stratospheric ozone depletion, ocean acidification, freshwater use, land-system use, introduction of novel entities and atmospheric aerosol loading. Identification and quantification of the control variables are often possible because of the effort by the international community to understand the planet's biogeochemical cycles and how these cycles have changed throughout Earth's history. In Steffen et al. (2015), control variables from seven of the nine boundaries have been quantified. SOLAS scientists have contributed to quantification of three of these boundaries: climate change, biogeochemical flows and ocean acidification. Indeed, in the early 2000s, SOLAS and the project IMBER (Integrated Marine Biogeochemistry and Ecosystem Research), in collaboration with IOCCP (International Ocean Carbon Coordination Project) brought together scientists with particular expertise to consider the specific research topic of ocean acidification. To facilitate the collaboration a working group was established, leading to the founding of the Ocean Acidification International Coordination Centre (OA-ICC) hosted by the International Atomic Energy Agency (IAEA). Steffen et al. (2015), using important outcomes from SOLAS research, have shown that in the domain of ocean acidification, humanity is still in the safe operating space defined by the authors, but is in the zone

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of uncertainty for climate change, and the high-risk zone for biogeochemical flows. This information is crucial for developing mitigation strategies and framing appropriate sustainability policy. Furthermore, the control variables of two of the nine planetary boundaries are not yet quantified due to a lack of understanding on atmospheric aerosol loading and introduction of novel entities (Steffen et al., 2015). In both these areas, ocean–atmosphere interface processes play a key role and so future research in the SOLAS realm will provide critical understanding of these two boundaries.

4. Future directions and challenges

Advancements in SOLAS research are required to assess the impact of anthropogenic activities on future climate and to inform policy relevant to ocean–atmosphere interactions. This progress can be achieved by a two-pronged approach of advances in science and integrated studies that inform policy decisions. Rapid changes in air–sea interactions are clearly occurring and it is critical that we continue to observe and understand these changes and eventually mitigate them. SOLAS science will continue to challenge our understanding of the Earth system, with the project's unique ability to facilitate essential integrated ocean–atmosphere research across scientific disciplines and national boundaries.

SOLAS (2015–2025) will address five core science areas: (1) greenhouse gases (GHG) and the oceans, (2) air–sea interface and fluxes of mass and energy, (3) atmospheric deposition and ocean biogeochemistry, (4) interconnections between aerosols, clouds, and marine ecosystems and (5) ocean biogeochemical control on atmospheric chemistry (http://www.solas-int.org/resources/books.html). While framing each of the five areas, the community has identified four requirements to achieve a step change in SOLAS science. Future efforts should consist of coordinated, integrated studies over large biogeographical regions and traditional disciplines, improvements in Earth system models, advances in remote sensing capabilities and in instrumentation and technique development, and access to remote platforms with continuous measurement capabilities.

All initial results of the last decade point to the need for more coordinated, large-scale, integrated studies. SOLAS is unique in providing a platform for bringing oceanographers and atmospheric scientists together, but in order to achieve more integrated studies, scientists with different types of expertise need to be engaged, such as modelers of large eddies, wave modelers, and biologists, for developing new observational techniques. Simultaneous studies of surface ocean plankton taxonomy/ecophysiology/bloom dynamics, surface concentrations of aerosol precursors and aerosol characteristics are required to constrain and model the biological and environmental drivers of biogenic aerosol emission. Time-series studies and inter-regional studies should be fundamental tools, as well as high quality measurements of the physical properties of the surface ocean mixed layer and the atmospheric MBL, to decouple the influence of ocean-derived aerosol on marine clouds from physical effects. Finally, it is recognized that in the complex, non-linear system of the surface ocean and lower atmosphere, the five SOLAS themes interact and influence each other. Understanding the processes involved, and generating projections, will not be possible by studying these themes independently. The community has identified a number of examples of regional oceanic systems where integrated studies are particularly urgent (Law et al., 2013), and need to be either initiated or expanded, including upwelling systems, sea ice areas, and coastal regions.

More complete Earth system models are an obvious tool for future SOLAS research. For climate projections on timescales of several hundred years, coupled Earth system models have been developed that include the most up to date knowledge on chemical and biogeochemical processes, but assimilation of data into biogeochemical ocean models is still in its infancy. With respect to marine aerosols, modeling should particularly address the variable stoichiometry of atmospheric nutrients and surface ocean biota, with better representations of competitive interactions between plankton groups, aerosols, and organic matter aggregation and export processes. Models of the biological and environmental drivers of biogenic aerosol emission as well as high-resolution numerical models to integrate cloud microphysics into small-scale process dynamics are urgently needed. Finally, atmospheric field experiments and associated modeling studies should be performed to understand the rates and pathways of atmospheric cycling of reactive emissions and how they interact with both the natural marine atmosphere and anthropogenic pollutants in continentally influenced regions.

Advances in remote sensing capabilities, instrumentation, and technique development will lead to greater process understanding in the next decade of SOLAS. Satellite observations of oceanic processes and atmospheric GHG concentrations have to be linked to oceanic measurements in a more systematic way. High-resolution satellite observations of aerosols, winds and cloud properties would help to improve process understanding and develop parameterizations of marine-cloud interactions. Recently, ground based instrumentation has been improved to make ocean-going measurements of fluxes of many trace gases feasible. Measurements of the exchange of a variety of volatile gases will help to identify and quantify transfer processes on different scales. Also, the sea-surface microlayer, which directly couples biological processes to atmosphere-ocean exchange, can now be probed remotely. Together with recent advances and techniques for research into small-scale interactions, this will undoubtedly lead to significant progress in our knowledge. New approaches for determining the emission flux of sea spray aerosols and secondary aerosol precursors, especially at high wind speeds would help to reduce uncertainties. Additionally, new techniques are needed for counting and characterizing nascent ultra-small aerosols to better assess the frequency and mechanisms of particle nucleation in the marine boundary layer.

Continuous measurement capabilities, especially those on remote platforms, are at the forefront of future SOLAS observational needs. Accurate, sustained observations and synthesis of greenhouse gases will be important in the next decade, especially with respect to the new technique of data-based surface ocean mapping (e.g., for CO_2, CH_4, and N_2O). Automated systems, such as high-accuracy pH sensors and alkalinity sensors, should be installed on profiling floats in order to monitor variablility in ocean acidification and their impacts. A 'Marine Atmospheric network' of coupled atmosphere-marine time-series sampling sites in both hemispheres, building on existing time series stations that monitor both atmosphere and ocean properties is also necessary. This network should utilize not only ships, but also buoys and island sites, and the temporal resolution of sampling at each site should be sufficient to resolve variability in both atmospheric deposition and ecosystem responses. These time series sites should also become focal points for detailed and in depth experiments and process studies. In addition, the impacts of ship plumes should explicitly be considered by evaluating how shipping traffic patterns are co-located with ocean–atmosphere observing sites. The long-term observation of the link between atmospheric material transport and marine biogeochemistry would facilitate both communication between groups working in different areas and development of universal parameterizations for implementation in numerical models.

For the past decade, SOLAS has demonstrated its interest and relevance to societal problems, for instance with respect to geoengineering schemes linked to the ocean–atmosphere system.

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Informed assessment of the feasibility, efficacy and potentially unintended effects of these schemes under debate for climate mitigation was derived from SOLAS science (e.g., Wallace et al., 2010). SOLAS will continue to provide the fundamental and essential knowledge to inform the geoengineering debate and policy makers on the critical aspects related to the interconnected ocean–atmosphere system. Furthermore, motivated by Future Earth, a high priority for SOLAS scientists in the upcoming decade will be to increase interaction with society and policy makers and to engage with researchers from the social science domains in order to expand the areas of SOLAS contribution beyond geo-engineering. New approaches will be investigated to launch projects meeting societal needs. Subjects addressed by SOLAS will include and focus more on climate regulation, evaluation of extreme weather events, cloud-aerosol interactions, carbon dioxide sequestration, air quality assessments, waste sinks and bioremediation, expansion of oceanic oxygen minimum zones, transport and accumulation of pollutants, and the fate of oil spills at the air–sea interface. SOLAS will assess the scope and structure of marine ecosystem services and contribute to the best possible use of nature-based solutions for sustainable development.

Over the past 10 years, SOLAS has made significant inroads regarding critical controls on the Earth system at the air–sea boundary. However, it is clear that this work has only scratched the surface of what we need to understand for our time in the Anthropocene. It cannot be ignored that there is a direct, two-way interaction between mankind and the air–sea system, and that both are undergoing unprecedented rates of change in the current epoch. The SOLAS community will address this challenge and continue the legacy of IGBP by studying more deeply the interactions between ocean and atmosphere in the Earth system science framework.

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